以週期性的空氣吹離法改善燃料電池一氧化碳毒化容忍度與微型燃料電池設計與性能分析

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摘 要

本論文分為兩部分。第一部分為運用不同週期的空氣吹離法改善被一氧化碳毒化之質子交換膜燃料電池性能。陽極燃料中如含有一氧化碳，一氧化碳會吸附白金觸媒造成燃料電池毒化現象產生，因此本論文研究當電池電位固定於 0.6 伏特時，以 4 個不同週期的空氣吹離法分別輸入含有不同濃度之一氧化碳的(20 ppm, 52.7 ppm 和 100 ppm)陽極燃料中觀測電池性能回復情形。由實驗結果可知，使用空氣吹離法可以明顯地增進燃料電池一氧化碳容忍度；除了一氧化碳濃度 100 ppm 外，當循環週期定為輸入空氣 10 秒接著關閉 10 秒時，改善電池性能的效果接近於持續地輸入空氣於被毒化的燃料電池中。

第二部分為研究設計微型質子交換膜燃料電池(反應面積為 2.5 cm^2)，微型燃料電池的流場板是使用微機電製程加工於矽晶圓材料上以蝕刻出流道，流場板表面沒有鍍金增加導電性，因此流場板並非如傳統燃料電池亦當雙極板使用，本論文改變習慣上組裝的順序，將電流收集板移至流場板

與氣體擴散層之間。本論文亦以模擬燃料電池電化學反應來觀測微型燃料電池內部質子交換膜電化學反應分布情形，電池內部的傳輸現象會遵守質量守恆、動量守恆、物種守恆、電荷守恆和能量守恆方程式，模擬以商業套裝軟體 CFD-ACE+完成，模擬的極化曲線與實驗的結果相互驗證，三維模型模擬結果分析兩個不同化學劑量比流量(3 和 5)的微型燃料電池，其觸媒層上電流密度、質子交換膜上水和溫度的分布情形。模擬結果發現微型燃料電池內部溫度分布受質子交換膜上氣態水分布所影響,模擬結果說明，當操作電壓定在 0.4 V 時,觸媒層上較低的化學劑量比流量會有較佳的電化學反應，因此會有較均勻的電流密度分布。另外，較低的化學劑量比流量會讓質子交換膜上水較多並均勻分布，造成較低且較均勻的溫度分布，模擬的結果可闡明微型燃料電池內部物理現象。

Improvement of CO Tolerance of PEMFC by Periodic Air Bleeding Technique & Design and Performance Analysis of micro PEMFC

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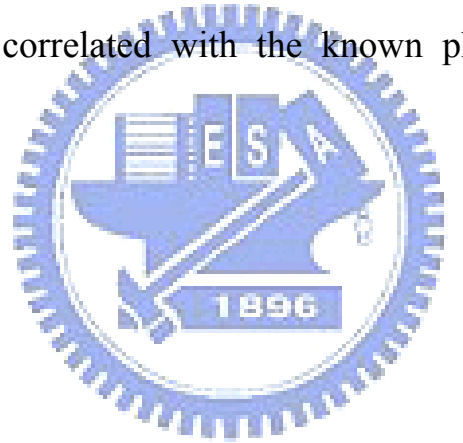
ABSTRACT

The dissertation consists of two subjects. Subject one investigates the improvement of carbon monoxide (CO) poisoning in a proton exchange membrane fuel cell (PEMFC) by using a periodic air dosing. The CO in the fuel gas leads to a significant loss in current density due to CO poisoning in the anode. The method of this experiment involves bleeding air into the anode fuel stream (H_2-CO), which contains CO in various concentrations (20 ppm, 52.7 ppm and 100 ppm). In the transient CO poisoning test, air bleeding is performed for four different periodic air dosing and cell voltage is fixed at 0.6 V. The results show that air bleeding technique can improve CO tolerance. The CO tolerance of a dosing air for 10 s in intervals of 10 s is similar to that of continuous air bleeding except 100 ppm CO. The CO tolerance of the fuel cell and cell performance recovery from poisoning can be improved by air bleeding.

Subject two designs a novel single micro proton exchange membrane fuel cell (active area is 2.5 cm^2) which is manufactured by applying Micro-electro-mechanical Systems (MEMS) technology on silicon substrates to etch flow channels without a gold-coating. A theoretical analysis is performed also in this study for such micro PEMFC. The three dimensional mathematical model transport phenomena in the micro PEMFC consists of the conservation equations of mass, momentum, energy, species and charge in a fully integrated

finite-volume solver using the CFD-ACE+ commercial code. The computed polarization curves are agreed well with experimental data. In addition, the predictions and analyses of micro PEMFC current density, water and temperature distributions in two different stoichiometric H_2/O_2 flows are given (3 and 5). The numerical results show that temperature distribution is affected by water distribution in the membrane. They also indicate that much more active electrochemical reaction in the catalyst layer at low flow rate leads to a superior current density distribution under 0.4 V operating voltage. In addition, a more and uniform water distribution induces a low and uniform temperature distribution in the membrane at low flow rate. Those illustrate that the model predictions are well correlated with the known phenomena of experimental mechanism.



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猶記當初碩士將畢業之際，在翁老師的鼓勵之下報考了交大博士班，口試時才知原來翁老師是陳教授剛回台灣任教時的第一位碩士班學生，因為這層徒孫的關係，讓我在口試時放下緊張忐忑的心，完整地敘述了碩士時的研究並也順利地錄取交通大學就讀博士班。光陰荏苒，我即將完成我博士的修業，在此，感謝翁老師在我碩士期間的啟蒙，讓我認識燃料電池並深入研究；感謝陳教授對我的栽培，讓我有獨立思考、判斷問題並解決問題的能力，希望陳教授在體能還能允許下，不要輕言退休，替國家作育更多英才；感謝交大給我機會讓我修讀博士學位，未來我必回饋母校來報答交大對我的期許；感謝所有愛我及我愛的人，沒有你們的容忍，我無法支持到今天，我愛你們！

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NOMENCLATURE

C	Reactant molar concentration (kmol/m^3)
C_{ref}	Reference molar concentration (kmol/m^3)
C_C^R	Reactant molar concentration of hydrogen in flow channel (kmol/m^3)
C_L^R	Reactant molar concentration of hydrogen in catalyst layer (kmol/m^3)
D_{eff}	Gas effective diffusivity (m^2/s)
D	Effective mass diffusion coefficient (m^2/s)
F	Faraday constant (C/kmol)
H_C	Flow channel height (m)
H_G	GDL height (m)
h	Gas enthalpy (J/kg)
i	Current density vector
J	Diffusive flux
J_C^{conv}	The mass flow rate from flow channel to GDL (kg/s)
J_G^{diff}	The mass flow rate from GDL to catalyst layer (kg/s)
j	Transfer current density (A/m^2)
j_e	Exchange current density (A/m^2)
M	Mixture molecular weight (kg/kmol)
N	Superficial flux (kmol/m^2·s)
n	Number of species, or number of electrons participating in a reaction
p	Absolute pressure (Pa)
q	Heat flux (J/m^2)
R	Universal gas constant (8.3143 kJ/kmol·K)
S	Reaction surface area (m^2)
Sh	Sherwood number

T	Absolute temperature (K)
U	Fluid velocity vector (m/s)
u	Thermodynamic equilibrium potential (V)
V	Medium volume (m^3)
v	Pore-water velocity in membrane (m/s)
x	Mole fraction
Y	Mass fraction
z	Charge number

Greek symbols

α	Mass transfer coefficient
β	kinetic constant
ε	Porosity
ρ	Fluid density (kg/m^3)
η	Over-potential (V)
κ	Permeability (m^2)
μ	Dynamic viscosity (kg/m·s)
σ	Electrical conductivity (1/Ω·m)
τ	Shear stress tensor (N/m^2)
δ	Tortuosity
φ	Concentration exponent
ώ	Mass production rate in gas phase ($kg/m^3 \cdot s$)
Φ	Potential (V)

Superscripts

conv	Convection

diff	Diffusion
K	Reaction kinetics
N	Nerst
R	Molar
tot	Total

Subscripts

an	Anode
C	Flow channel
ca	Cathode
con	Concentration loss
e	Exchange
eff	Effective value
G	Gas diffusion layer
in	Inlet
L	Catalyst layer